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Plastic scintillator for pulse shape neutrons and gamma quanta discrimination

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HIGHLIGHTS

- Plastic scintillator for efficient n/γ discrimination is described.
- Pulse shape discrimination is occurred due to direct transformation of triplet excitation energy.
- The scintillator contains two activation centers with different lifetimes.
- Eu-containing complexes were used as activators.
- With 3.0 wt% of Eu[DBM]$_3$Phen, this PS provides reliable discrimination with FOM = 1.37.

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ABSTRACT

This study describes a new plastic scintillator for pulse shape n-γ discrimination. The scintillator contains two activation centers with different life spans. The first activator collects the singlet excitation energy of a polymer base, and the second activator utilizes triplet excitation states. We utilized 1,4-dimethyl-9,10-diphenylanthracene (DMDPA) and tris(dibenzoylmethide) (1,10-phenanthroline)Europium(III) (Eu[DBM]$_3$Phen) as activators. The figure of merit for this scintillator is 1.37, which is sufficient for reliable n-γ discrimination.

1. Introduction

Pulse shape discrimination (PSD) is an important method used for the registration of fast neutrons in a gamma radiation background. This method is based on the radioluminescence pulse of most scintillators, which consists of fast (prompt) and slow (delayed) components. The major part of a scintillating light carries the fast singlet component and has a lifetime of less than 10 ns. The lifetime of the slow triplet component, whose intensity increases with the ionization density of detected particles is approximately 2 μs. The ratio of fast and slow component intensities can be a criterion for identifying particles (including n-γ discrimination) (Birks, 1964).

The best and most frequently used organic scintillators for both pulse shape neutron and gamma discrimination are organic monocrystals such as stilbene, anthracene, *p*-terphenyl, and others (Birks, 1964; Daenhick and Sherr, 1961). The typical value of the stilbene n-γ discrimination figure of merit (FOM)[1] is 4.7 (Zaitseva et al., 2012). For statistically significant PSD, the FOM must be greater than 1.27, which corresponds to a 3σ error (Zaitseva et al., 2012).

Liquid scintillators NE-213, NE-230, NE-232 and their analogs are also widely used for n-γ discrimination. However, their FOM ($\sim$3.1) is less than that of organic single crystals (Zaitseva et al., 2012; Peuckert, 1962; Bovet et al., 1972).

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[1] FOM = $S/(\delta_{gamma} + \delta_{neutron})$, where S is the separation between gamma and neutron peaks, and δ_{gamma} and $\delta_{neutron}$ are the full widths at half maximum of the corresponding peaks (Zaitseva et al., 2009).

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Despite such high PSD characteristics, organic crystal scintillators have drawbacks that significantly limit their use including small size, high cost, and complicated growing and machining processes. Liquid scintillators have a low fire point and are more difficult to use than solid detectors. Plastic scintillators (PSs) have no such drawbacks, but there are a limited number of studies investigating their use for n-γ PSD.

Brooks et al. (1960) presented a polyvinyltoluene-based PS with isopropyldiphenyl. This PS, which they called "plastic 77", contains *p*-terphenyl (p-TP) as a first luminescent additive and 1,4-bi-(2-(5-phenyloxazolile))-benzene (POPOP) as a second luminescent additive. According to Brooks and Pringle, "plastic 77" can be used for *n*-γ discrimination. Unfortunately, they did not present any quantitative parameters of *n*-γ discrimination quality.

A polystyrene-based PS with isopropyldiphenyl was studied by Grudskaya et al. (1968). The discrimination ability of a PS for α- and β-particles is 1.18–1.21. Grudskaya et al. qualitatively confirmed the possibility of using PS for *n*-γ discrimination because it follows from their presented graphical data. However, the quantitative characteristics of *n*-γ discrimination were not presented.

Zaitseva et al. (2012) described the *n*-γ discrimination of a polyvinyltoluene-based PS with a high concentration (up to 37%) of 2,5-diphenyloxazole (PPO). This PS provides an FOM of 3.31, which is close to that of the commercially available liquid scintillator EJ-301 (FOM = 3.21). The high value of *n*-γ discrimination parameters was obtained because of the high PPO concentration. The high concentration significantly increases the rate of the triplet–triplet annihilation and increases the part of the slow component of a scintillator response. The high concentration of the additive close to its solubility limit decreases the transparency (because of reabsorption), mechanical strength, and long-term stability of the PS. Additionally, the high concentration limits its dimensions and application fields and makes the cost similar to that of organic single crystals.

All known PSs for *n*-γ discrimination have significant drawbacks that impair their industrial production. It is possible this is a reason for the lack of commercially available PSs for PSD. Nevertheless, the new type of plastic scintillator by Eljen Technology named EJ-299-33 have to be noted (Pozzi et al., 2013).

All PSs for *n*-γ discrimination use the presence of fast and slow components in a scintillating response. These components are formed in processes of singlet-level deactivation (fluorescence) and triplet–triplet annihilation. It is the triplet component that carries information about a particle type because it is sensitive to the stopping power (*dE/dx*) of the particle (Birks, 1964).

In this study, we propose another PS for *n*-γ discrimination. In our PS, the triplet energy transformation is directly performed by the specific activators. Therefore, this system does not require triplet–triplet annihilation. As a result, it is possible to decrease the concentration of activators by an order of magnitude while simultaneously improving the optical and mechanical properties of the scintillator.

2. Experimental

2.1. Choosing the singlet and triplet energy activators

In this study, we chose to use a tris(dibenzoylmethide)(1,10-phenanthroline) europium(III) (Eu[DBM]$_3$Phen) complex as an efficient activator of triplet excitations for further investigation. The complex was used previously as a component of an efficient alpha particle detector (Adadurov et al., 2010, 2011). During irradiation, the triplet excitation energy of a polymer base is transferred to the Eu^{3+} ion, which emits energy at a 612 nm wavelength with a 370 μs lifetime. The singlet energy is also transferred to the complex, but its contribution is less than 25% of the total because the population of triplet levels is three times higher than that of the singlet levels (Birks, 1964). Thus, the Eu[DBM]$_3$Phen complex appears to be suitable for the slow component of the scintillating pulse formation.

For the singlet energy transformation and the fast component formation, it is clear that the same activators that are used in common commercial scintillators should be used. However, our measurements have shown that the scintillating response of a polystyrene-based PS with 2% wt p-TP and 2% wt Eu[DBM]$_3$Phen lacks the fast component corresponding to p-TP fluorescence. This result indicates that the Eu[DBM]$_3$Phen complex completely quenches the fluorescence. An analogous effect was also observed for other known activators such as PPO, PBD, etc. To understand the causes of quenching, we measured the excitation and fluorescence spectra of two polystyrene films with 2% wt p-TP and 2% wt Eu[DBM]$_3$Phen, respectively. Measurements were made using a Fluoromax-4 (HORIBA Jobin Yvon Inc., Edison, NJ) spectrofluorometer. The measured spectra are presented in Fig. 1. The data indicate that the p-TP fluorescence spectrum is entirely overlapping the Eu[DBM]$_3$Phen absorption spectrum, which explains the observed quenching.

To solve the problem of *n*-γ discrimination, it is necessary to find an additive with an absorption that overlaps the polystyrene emission but with an emission that lies out of the Eu[DBM]$_3$Phen absorption range. This requirement is satisfied by 1,4-dimethyl-9,10-diphenylanthracene (DMDPA). The structural formula of this compound is presented in Fig. 2.

The DMDPA molecule contains two types of chromophore centers: a dimethylanthracene core and two phenyl rings in the 1 and 4 positions. This provides a large Stokes shift of this compound because absorption is determined by phenyl chromophores while fluorescence is determined by the massive dimethylanthracene core.

This compound was prepared according to the method described by Krasovitskii and Afanasiadi (1977), with additional purification. The excitation and fluorescence spectra of a polystyrene film with 1.5% wt DMDPA are presented in Fig. 3. The fluorescence was excited at a wavelength of 265 nm (the polystyrene fluorescence maximum). The excitation spectrum was

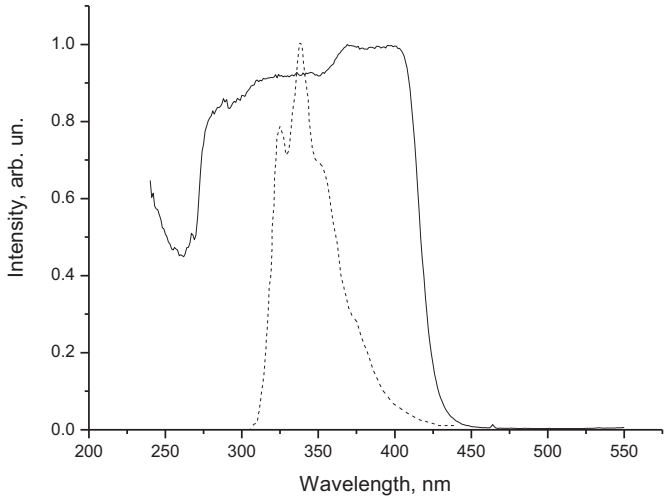

Fig. 1. Excitation spectrum of polystyrene film with 2% wt of Eu[DBM]$_3$Phen complex (solid line) and fluorescence spectrum of polystyrene film with 2 wt% of p-TP (dashed line). The observation wavelength was 612 nm, and the excitation wavelength was 265 nm.